

Thermal Stability of InGaN/GaN Multiple Quantum Wells Grown by Metalorganic Chemical Vapor Deposition

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Recently, InGaN alloys have attracted much attention for the realization of blue-green light-emitting diodes and laser diodes. Due to the solid phase immiscibility of InN and GaN, phase separation was reported previously in thick InGaN films and InGaN/GaN multiple quantum wells (MQWs) upon thermal annealing. Interdiffusion of In and Ga in $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}/\text{GaN}$ MQW structures was observed at annealing temperature above the theoretically critical temperature. Additionally, indium-outdiffusion during the epitaxial growth was also proposed. In this work, an in-depth investigation of the interdiffusion of InGaN heterojunctions is performed to study their thermal stability.

In this study, $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ multiple quantum wells (MQWs) with InN mole fractions of ~ 0.23 and ~ 0.30 were grown by low-pressure metalorganic chemical vapor deposition on c-plane sapphire substrates. These samples were then subject to postgrowth thermal annealing at various temperatures and duration. The optical properties were assessed by low temperature photoluminescence (PL) measurements to examine the interdiffusion in these samples due to thermal annealing. To extract the diffusion coefficient, Fick's law was used to calculate the indium concentration profile after annealing and self-consistent calculation was performed including the effect of free-carrier screening on the polarization fields in wurtzite InGaN/GaN structures. Parabolic energy dispersion relation was used in the calculation.

Experimental results show that the integrated PL intensity initially increases and then decreases with annealing time for the samples annealed above 1000°C . A maximum PL intensity can be obtained after annealing at 1000°C for 20 minutes. However, no much enhancement of PL intensity is achieved for the samples annealed at low temperature. In addition, a blue shift of PL peak energy is induced by thermal annealing at $900\text{--}1050^\circ\text{C}$ for 10 minutes. This is attributed to the decrease of indium concentration in the quantum well after the intermixing between InGaN quantum wells and GaN barriers. A large blue shift of about 30 meV and degradation of optical quality for the sample annealed at 1050°C for 10 minutes was observed. This implies that there is a thermal budget issue for the later growth since severe interdiffusion of In and Ga may occur at the typical growth temperature of GaN.

The diffusion coefficients at different annealing temperatures were extracted by a least-squares fit to the square of the diffusion length against annealing time. The deduced diffusion coefficient varies from 2.1×10^{-20} to $1.3 \times 10^{-18} \text{ cm}^2/\text{sec}$ as the annealing temperature varies from 900 to 1050°C for $\text{In}_{0.23}\text{Ga}_{0.77}\text{N}/\text{GaN}$ MQWs. For $\text{In}_{0.31}\text{Ga}_{0.69}\text{N}/\text{GaN}$ MQWs, the diffusion coefficients are in the range of 1.2×10^{-19} to $7.7 \times 10^{-18} \text{ cm}^2/\text{sec}$, which are comparable to the reported data. The activation energy for the interdiffusion is calculated to be $3.4 \pm 0.5 \text{ eV}$ with the prefactors of 1.6×10^{-4} to $2.5 \times 10^{-4} \text{ cm}^2/\text{sec}$ for $\text{In}_{0.23}\text{Ga}_{0.77}\text{N}/\text{GaN}$ and $\text{In}_{0.31}\text{Ga}_{0.69}\text{N}/\text{GaN}$ MQWs, respectively. Compared to InGaAs/GaAs and AlGaAs/GaAs material systems, we propose that the interdiffusion in InGaN/GaN should be governed by a single mechanism, i.e. vacancy-controlled second-nearest-neighbor hopping.

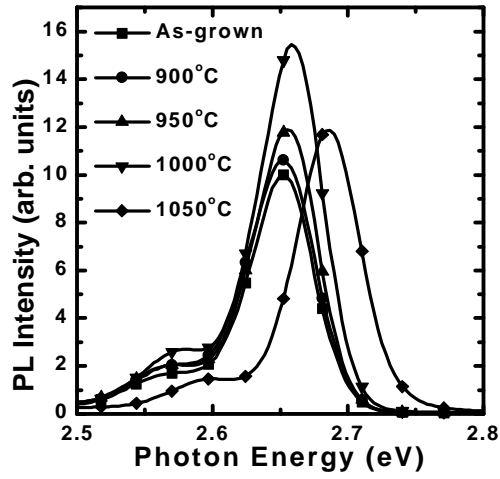


Fig. 1 25K photoluminescence spectra of the as-grown and annealed $\text{In}_{0.23}\text{Ga}_{0.77}\text{N}/\text{GaN}$ MQWs.

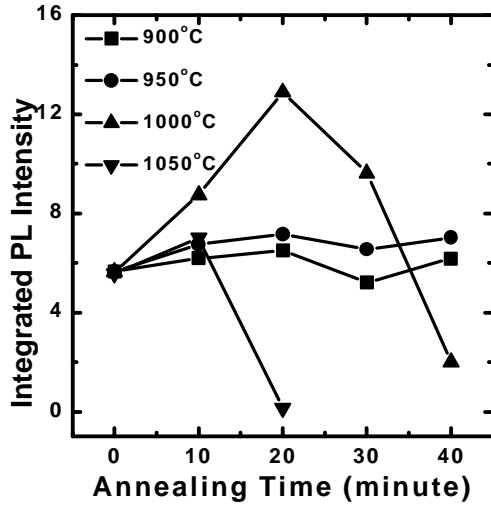


Fig. 2 Integrated photoluminescence intensity as a function of thermal annealing time at different annealing temperatures.

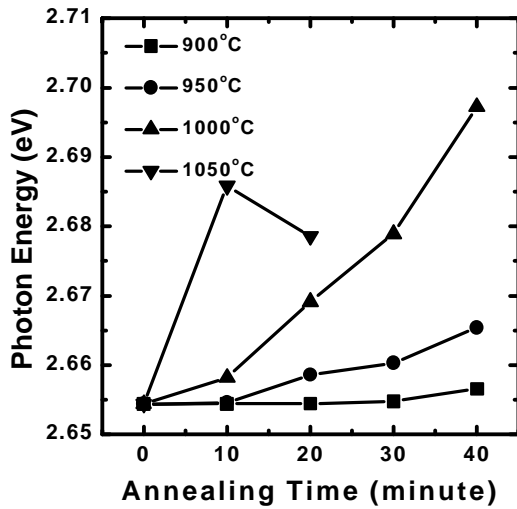


Fig. 3 Variation of PL peak energy with annealing time at different annealing temperatures.

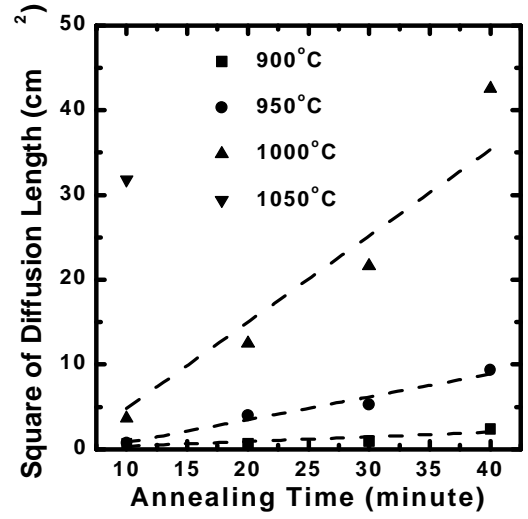


Fig. 4 The square of diffusion length versus annealing time at different temperatures.

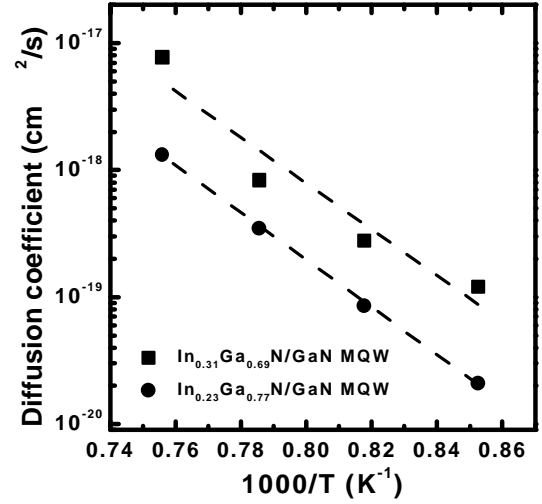


Fig. 5 Arrhenius plot of $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ interdiffusion coefficients for temperatures between 900 °C and 1050 °C.